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# Dispersion-engineered and highly-nonlinear microstructured polymer optical fibres

Michael H. Frosz<sup>a</sup>, Kristian Nielsen<sup>a</sup>, Petr Hlubina<sup>b</sup>, Alessio Stefani<sup>a</sup>, and Ole Bang<sup>a</sup>

<sup>a</sup>DTU Fotonik, Department of Photonics Engineering, Technical University of Denmark, Ørstedes Plads 343, DK-2800 Kgs. Lyngby, Denmark;

<sup>b</sup>Department of Physics, Technical University of Ostrava, 17. listopadu 15, 708 33 Ostrava-Poruba, Czech Republic

## ABSTRACT

We demonstrate dispersion-engineering of microstructured polymer optical fibres (mPOFs) made of poly(methyl methacrylate) (PMMA). A significant shift of the total dispersion from the material dispersion is confirmed through measurement of the mPOF dispersion using white-light spectral interferometry. The influence of strong loss peaks on the dispersion (through the Kramers-Kronig relations) is investigated theoretically. It is found that the strong loss peaks of PMMA above 1100 nm can significantly modify the dispersion, while the losses below 1100 nm only modify the dispersion slightly. To increase the nonlinearity of the mPOFs we investigated doping of PMMA with the highly-nonlinear dye Disperse Red 1. Both doping of a PMMA cane and direct doping of a PMMA mPOF was performed.

**Keywords:** Microstructured polymer optical fibre (mPOF), dispersion engineering, influence of loss on dispersion, dye doping, poly(methyl methacrylate) (PMMA), Disperse Red 1

## 1. INTRODUCTION

The huge interest in silica photonic crystal fibres (PCFs) has largely been due to the possibility of manipulating the dispersion profile by modifying the microstructure. This has allowed researchers to shift the zero-dispersion wavelength of silica fibres to below 800 nm by reducing the core size.<sup>1</sup> The combination of a small core size and zero-dispersion wavelength at the operating wavelength of widely available femtosecond Ti:sapphire lasers led to an extensive research in supercontinuum generation and other nonlinear effects in PCFs.<sup>2</sup> It is crucial for the efficiency of many nonlinear mechanisms that the pump laser wavelength is close to the zero-dispersion wavelength and that the core size is small.

Recently, work in fabricating PCFs from materials other than silica, such as chalcogenide glasses<sup>3</sup> or polymer,<sup>4,5</sup> has intensified. One of the advantages of using alternative materials can be extended wavelength transmission range or a higher inherent material nonlinearity. Depending on the type of polymer used, microstructured polymer optical fibres (mPOFs) can potentially be made highly-nonlinear. Another advantage is that polymer materials have a higher bio-compatibility than silica, meaning that it is easier to bond certain types of biosensor materials to a polymer surface than to silica.<sup>6</sup>

We have fabricated mPOFs in the polymer poly(methyl methacrylate) (PMMA) with core sizes down to about 2-3  $\mu\text{m}$  diameter using a simple two-step process. The structured PMMA fibre preform is first drawn into a cane, reducing the structure by a factor of  $\sim 10$ . The cane is then inserted into a PMMA tube and drawn into a fibre, reducing the structure by an additional factor of  $\sim 30$ .

We present a characterisation of our small-core mPOFs, including scanning electron microscope (SEM) imaging of the structure, loss measurements and dispersion measurements. The dispersion measurements show that we have successfully obtained significant modification of the dispersion compared to the dispersion of the bulk material. We then theoretically investigate the influence of the high material losses on the dispersion of mPOFs made of PMMA. Finally, we demonstrate solution doping of PMMA with the highly nonlinear dye Disperse Red

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Further author information:

M.H.F.: E-mail: mhfr@fotonik.dtu.dk, Telephone: +45 45 25 63 69

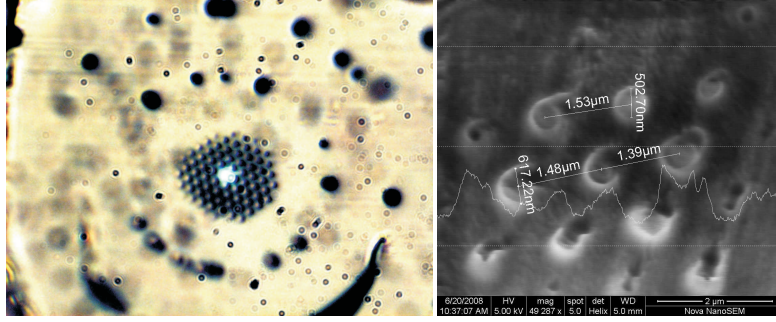


Figure 1. *Left:* Optical microscope image of an mPOF guiding light at 1064 nm. It is clearly seen how the light is confined to the core within the microstructure. *Right:* Scanning electron microscope image of an mPOF with a pitch  $\Lambda$  of  $\sim 1.5 \mu\text{m}$  and relative hole size  $d/\Lambda \sim 0.4$ . The core diameter is  $\sim 2.5 \mu\text{m}$ .

1 (DR1) using two approaches. First, we have doped a cane and afterwards drawn it into a fibre, as demonstrated by Large *et al.* who used the dye Rhodamine 6G.<sup>7</sup> Second, we successfully doped an mPOF directly by placing the fibre in a solution containing the nonlinear dye.

## 2. DISPERSION-ENGINEERING OF MICROSTRUCTURED POLYMER OPTICAL FIBRE

Images of PMMA mPOFs are shown in Fig. 1. It is seen how it is possible for us to fabricate mPOFs with sub-micron air-holes while the core guides light. Because the holes can be made so small, they can be difficult to see in an optical microscope and it can be necessary to use SEM-imaging to measure the hole size.

By making the core small enough it is possible to engineer a waveguide-dispersion contribution large enough to significantly shift the total dispersion profile of an mPOF. To find the structural parameters (pitch  $\Lambda$  and relative hole size  $d/\Lambda$ ) necessary to obtain a particular zero-dispersion wavelength, we calculated the dispersion for a wide range of structural parameters. The dispersion calculations were made using a mode solver based on the finite element method.<sup>8</sup> The material dispersion of PMMA was included using

$$n^2(\lambda) = A_0 + A_1\lambda^2 + A_2\lambda^{-2} + A_3\lambda^{-4} + A_4\lambda^{-6} + A_5\lambda^{-8} \quad (1)$$

where  $A_0 = 2.18645820$ ,  $A_1 = -2.4475348 \times 10^{-4} \mu\text{m}^{-2}$ ,  $A_2 = 1.4155787 \times 10^{-2} \mu\text{m}^2$ ,  $A_3 = -4.4329781 \times 10^{-4} \mu\text{m}^4$ ,  $A_4 = 7.7664259 \times 10^{-5} \mu\text{m}^6$ , and  $A_5 = -2.9936382 \times 10^{-6} \mu\text{m}^8$ . This expression was found as a fit to the measured refractive index in the range 365–1060 nm for the same type of PMMA as we use<sup>9</sup> (the optical properties of PMMA vary depending on the fabrication method). The results are shown in Fig. 2 where the zero-dispersion wavelength (vertical axis) is shown as a function of both pitch  $\Lambda$  (horizontal axis) and relative hole size  $d/\Lambda$  (colourscale). Using the figure one can therefore determine the structural parameters necessary to obtain a given zero-dispersion wavelength. If, e.g., a zero-dispersion wavelength of 1064 nm is desired, one can find from the figure that a pitch  $\Lambda = 2.1 \mu\text{m}$  and relative hole size  $d/\Lambda = 0.6$  results in the target zero-dispersion wavelength of 1064 nm.

To demonstrate the possibility of modifying the total dispersion by engineering a large waveguide dispersion, we fabricated two mPOFs. The first mPOF, termed “LMA” for Large Mode Area, was made with a pitch  $\Lambda = 11 \mu\text{m}$ , and  $d/\Lambda = 0.36$ , so that the core is relatively large ( $\sim 18 \mu\text{m}$ ). The total dispersion is then expected to be dominated by material dispersion. The second mPOF was made with a relatively small core of  $3.8 \mu\text{m}$  ( $\Lambda = 2.4 \mu\text{m}$ ,  $d/\Lambda = 0.4$ ) and termed “DSF” for Dispersion Shifted Fibre; in this case the waveguide dispersion is expected to shift the total dispersion significantly away from the material dispersion. The dispersion of the fabricated mPOFs was measured using white-light spectral interferometry<sup>10,11</sup> and is shown in Fig. 3. It is seen from the figure that the measured dispersion of the LMA fibre is very close to the material dispersion of PMMA, as expected. It is also seen how proper design of the microstructure in the DSF fibre can significantly shift the dispersion away from the material dispersion. This demonstrates that the extremely useful dispersion-engineering of silica PCFs<sup>1</sup> is also possible in PMMA mPOFs.

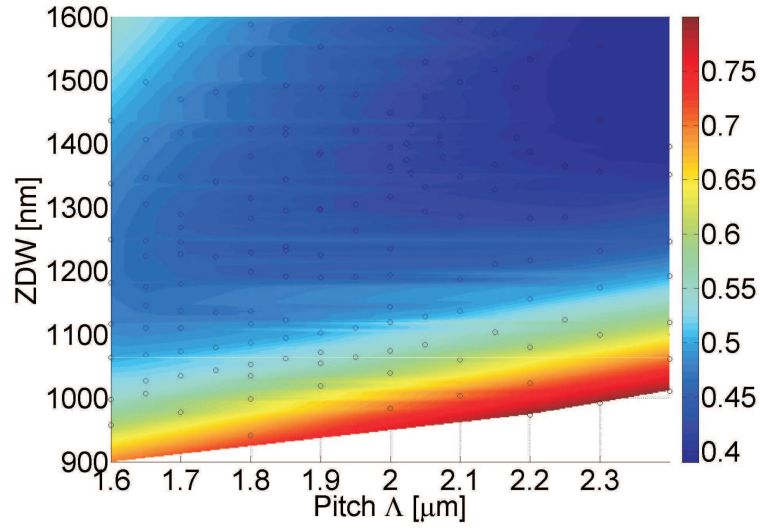


Figure 2. The zero-dispersion wavelength of a triangularly structured mPOF made of PMMA, shown as a function of both pitch  $\Lambda$  (horizontal axis) and relative hole size  $d/\Lambda$  (colorscale). Colour available in digital version.

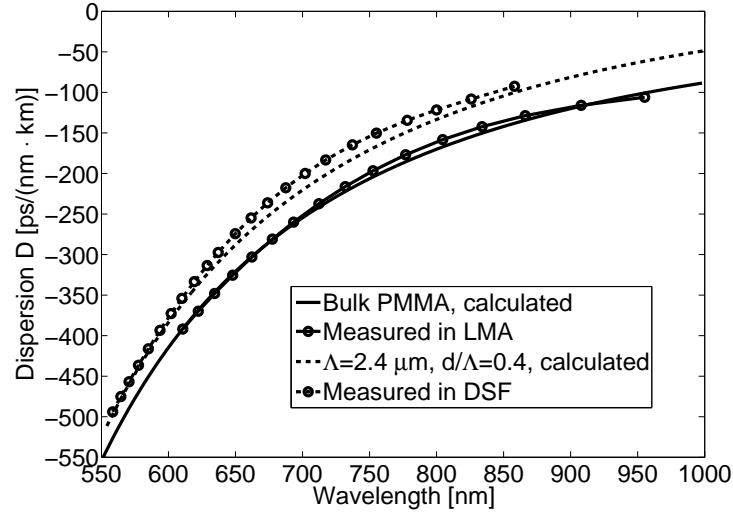


Figure 3. Comparison of measured and calculated dispersion in PMMA mPOF with and without significant waveguide contribution. Solid line: calculated dispersion of bulk PMMA; solid line with circles: measured dispersion of LMA mPOF with relatively large core. Dashed lines: calculated (no circles) and measured (with circles) dispersion of PMMA dispersion-shifted mPOF with  $\Lambda = 2.4 \mu\text{m}$ ,  $d/\Lambda = 0.4$ .

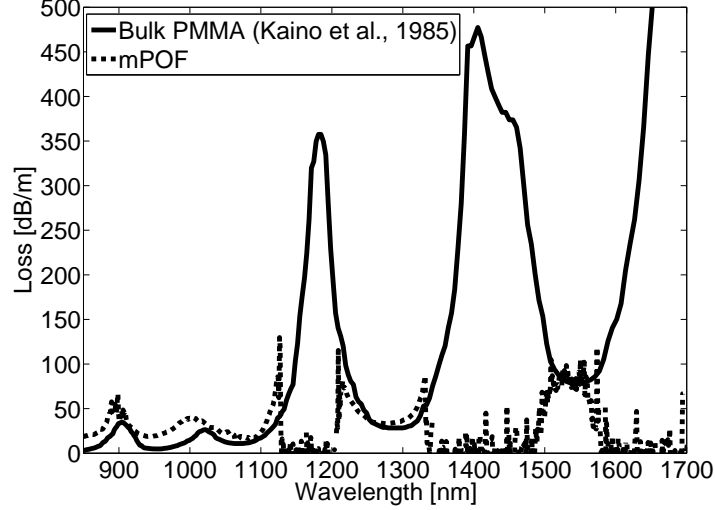


Figure 4. The measured loss in bulk PMMA from Ref. 12 (solid line) and the measured loss in an mPOF (dashed). The losses for the mPOF fibre could not be measured in wavelength regions with very high loss (around 1180 nm, 1400 nm, and above 1600 nm).

### 3. INFLUENCE OF LOSS PEAKS ON DISPERSION

The optical loss spectrum of PMMA exhibits some very significant peaks in the near-infrared due to harmonics of the vibrational C-H bond.<sup>12</sup> The measured loss in bulk PMMA is shown in Fig. 4 together with the measured loss for an mPOF. It is clear that material losses are dominant in the mPOF fibre.

Since the loss is related to the refractive index of a material through the Kramers-Kronig relations, these loss peaks could potentially significantly modify the material dispersion in the vicinity of the loss peaks. To investigate this we used the measured loss of bulk PMMA from Ref. 12. The change in the imaginary part  $\chi''$  of the susceptibility  $\chi$  due to the loss  $\alpha$  is found from<sup>13</sup>

$$\alpha(\nu) = -\frac{2\pi\nu}{n_0 c_0} \chi''(\nu), \quad \chi''(\nu) \ll 1. \quad (2)$$

where  $n_0$  is the refractive index in the absence of the loss resonances and  $c_0$  is the speed of light in vacuum. The change in the real part  $\chi'$  of the susceptibility is then found from the Kramers-Kronig relation<sup>13</sup>

$$\chi'(\nu) = \frac{2}{\pi} \int_0^\infty \frac{s \chi''(s)}{s^2 - \nu^2} ds, \quad (3)$$

and finally the change in refractive index due to the loss is found from<sup>13</sup>

$$n(\nu) - n_0 = \frac{\chi'(\nu)}{2n_0}, \quad \chi'(\nu) \ll 1. \quad (4)$$

For  $n_0$  we use the refractive index given by Eqn. (1). The calculated change in refractive index due to the bulk loss of PMMA is shown in Fig. 5. It is seen that the change in refractive index  $n$  is on the order of  $10^{-5}$ , so initially one could think that the influence of loss on dispersion is negligible. However, one must keep in mind that the change of the slope of  $n$  with frequency  $\nu$  is highly important for the dispersion. We have therefore calculated the dispersion for an mPOF both using the material dispersion in the absence of loss and the calculated material dispersion in the presence of loss. The result is shown in Fig. 6. It is seen that when the change in material dispersion due to loss is included, the dispersion can be strongly modified in the vicinity of the loss peaks. This is most clear around the loss peaks at  $\sim 1180$  nm and  $\sim 1400$  nm within the wavelength range shown in the

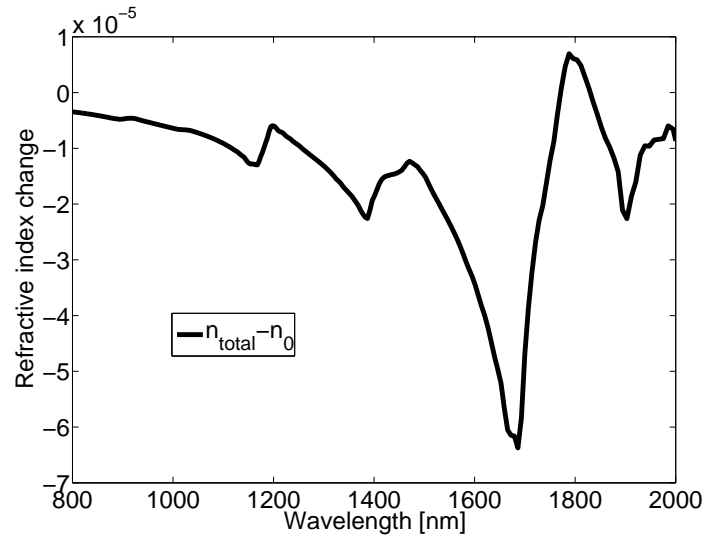


Figure 5. The calculated change in refractive index due to loss in PMMA.

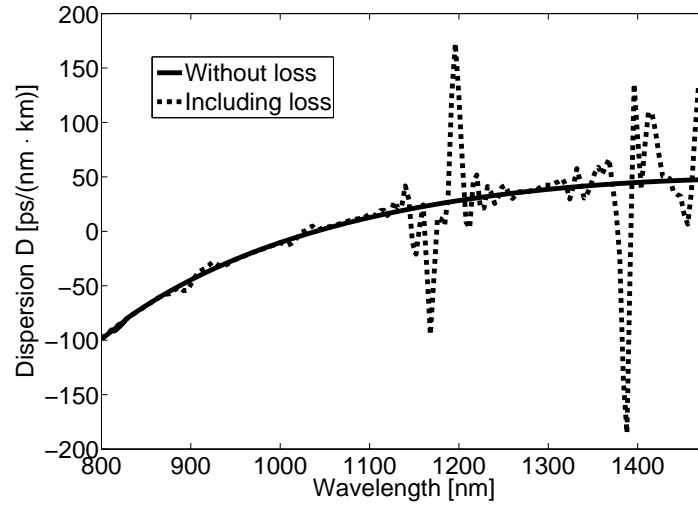


Figure 6. The calculated dispersion for an mPOF with  $\Lambda = 2.0 \mu\text{m}$  and  $d/\Lambda = 0.6$  when only the material dispersion in absence of loss is taken into account (solid line) and when the influence of loss on the refractive index is included (dashed line).

figure. For the loss peak around  $\sim 1020$  nm of about 26 dB/m, the shift in dispersion due to material loss is only about  $\pm 5$  ps/(nm · km). The change in dispersion due to material loss could, nevertheless, still play a role for nonlinear optical effects, where even a small change in dispersion can lead to a significant change of the output spectrum.<sup>14</sup>

#### 4. INCREASING THE NONLINEARITY BY DOPING

Although mPOFs can be fabricated with the same core size as used in highly nonlinear silica PCFs,<sup>15</sup> it is still necessary for the nonlinearity of the polymer material to be sufficiently high to overcome the detrimental effects of the very high transmission losses of typical polymers such as PMMA compared to the low losses obtained in silica. To estimate the nonlinearity of neat PMMA and compare it to silica we use the nonlinear-index coefficient  $n_2 \propto \text{Re}(\chi^{(3)})/n$ , where  $\chi^{(3)}$  is the third-order susceptibility and  $n$  is the refractive index.<sup>16</sup> Using the values stated in Refs. 17 and 18 at  $\lambda = 1064$  nm we obtain

$$\frac{n_{2,\text{PMMA}}}{n_{2,\text{silica}}} = \frac{\chi_{\text{PMMA}}^{(3)}}{\chi_{\text{silica}}^{(3)}} \cdot \frac{n_{\text{silica}}}{n_{\text{PMMA}}} = \frac{7 \times 10^{-14} \text{ esu}}{3.1 \times 10^{-14} \text{ esu}} \cdot \frac{1.44967}{1.4795} \approx 2. \quad (5)$$

We can thus expect the nonlinearity of neat PMMA to be approximately twice that of silica. However, since the loss  $\alpha$  at 1064 nm in PMMA is approximately  $10^4$  times larger than in silica, the figure of merit  $n_2/\alpha$  for PMMA is  $\sim 2 \times 10^{-4}$  that of silica.

We have therefore considered how to increase the nonlinearity of mPOFs. This has been achieved previously for step-index polymer fibres by adding a highly nonlinear dye to the polymer preform before drawing it into a fibre.<sup>19,20</sup> The dye was added to the monomer during polymerisation. The chemical processes required for the polymerisation can be avoided by instead solution doping the polymer. This was recently demonstrated by drawing a neat PMMA preform into a cane, and then placing the cane in a solution containing the dye Rhodamin 6G.<sup>7</sup> The dye then diffuses into the polymer thereby doping it. When the doping is complete the cane is drawn into an mPOF.

We used a similar procedure for doping a PMMA cane but used the highly-nonlinear dye Disperse Red 1 (DR1). To estimate the nonlinearity of DR1 we again use the values stated in Refs. 17 at  $\lambda = 1064$  nm to obtain

$$\frac{n_{2,10\% \text{DR1/PMMA}}}{n_{2,\text{silica}}} \approx \frac{\chi_{10\% \text{DR1/PMMA}}^{(3)}}{\chi_{\text{silica}}^{(3)}} \cdot \frac{n_{\text{silica}}}{n_{\text{PMMA}}} = \frac{141 \times 10^{-14} \text{ esu}}{3.1 \times 10^{-14} \text{ esu}} \cdot \frac{1.44967}{1.4795} \approx 45, \quad (6)$$

where we neglect the difference between  $n_{\text{PMMA}}$  and  $n_{10\% \text{DR1/PMMA}}$ . The nonlinearity of a 10% solution of DR1 in PMMA is thus expected to be approximately 45 times larger than in silica.

First, DR1 in the form of powder was dissolved in a 50/50 mixture of ethylene glycol and methanol. The ethylene glycol is used to dissolve the DR1 powder and the methanol is used to increase the diffusion of the dye into the PMMA.<sup>21</sup> The PMMA cane is then placed in a container with the solution containing DR1 and ethylene glycol/methanol so that the solution fills the air-holes of the cane, and the dye can then diffuse into the core. After about two days the cane is removed from the solution and residual methanol is removed by heating the cane to 80 °C. A microscope image of the end face of the cane is shown in Fig. 7. It is seen how some of the holes have been clogged by undissolved dye powder, while other holes have not been clogged and only contain air. This has led to a non-uniform doping of the cane, which can be avoided by more thorough dissolution of the dye powder and/or a lower concentration of dye in the solution.

As an alternative method of doping we also tried taking an mPOF drawn from neat PMMA and placing it in a solution containing DR1 and ethylene glycol/methanol. By keeping the fibre ends out of the solution we ensured that any undissolved powder could not clog the air-holes. The dye therefore diffused into the fibre through the outer surface. A microscope image of the end face of the mPOF after doping is shown in Fig. 8. It is seen how the doping appears to be uniform over the entire end face of the mPOF. This direct fibre doping method therefore seems promising for doping fibres made of neat PMMA with a highly-nonlinear dye. The method has the further advantage that it is also possible to measure the transmission, using an optical spectrum analyser



Figure 7. An optical microscope image of the end face of the cane doped with DR1. Colour available in digital version.

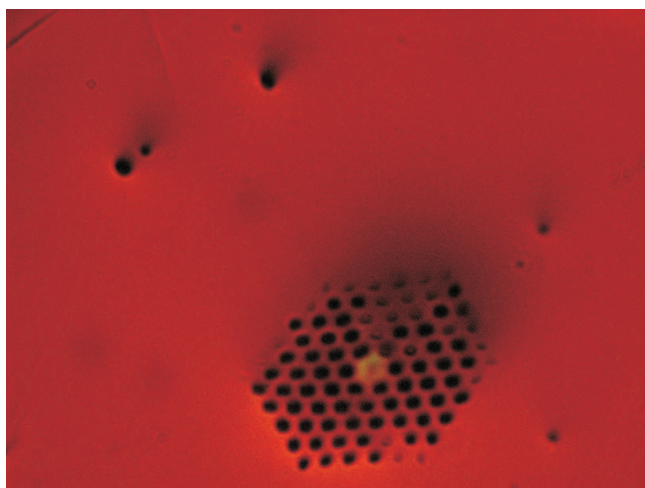


Figure 8. An optical microscope image of the end face of the mPOF directly doped with DR1. Colour available in digital version.



and a broadband light source, in real-time during doping. One can therefore study the diffusion process over time and accurately control the dopant concentration in the core.

Finally, we note that the doping is naturally expected to change the dispersion of the mPOF. It is possible to calculate the refractive index of PMMA doped with DR1 as a function of wavelength<sup>22</sup> and therefore also to calculate the dispersion of mPOFs doped with DR1. Dispersion engineering will therefore also be possible in these highly-nonlinear doped fibres.

## 5. CONCLUSIONS

We have demonstrated dispersion-engineering of microstructured polymer optical fibres (mPOFs) by drawing an mPOF with a relatively small core, and measuring its dispersion to be significantly shifted from the material dispersion of PMMA. This means that it is possible to draw mPOFs with a zero-dispersion wavelength optimised for a particular pump wavelength, thereby enhancing the efficiency of nonlinear effects.

We also investigated the influence of the strong loss peaks of PMMA on the material dispersion and found that the dispersion is modified quite significantly in the vicinity of the strongest loss peaks ( $> 100$  dB/m). For the loss peak of about 26 dB/m at  $\sim 1020$  nm the dispersion was only modified by about  $\pm 5$  ps/(nm · km).

Finally, we described doping of a cane with the highly-nonlinear dye Disperse Red 1 (DR1). Further, we found that direct doping of an mPOF made from neat PMMA is also possible, thereby avoiding potential problems with clogging of the air-holes in the cane with undissolved dye powder. This latter method has the additional advantage that it is possible to measure the progress of diffusion doping in real-time by making a simple transmission measurement.

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